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Der Präsident des Europäischen Patentamts;
Im Auftrag

For the President of the European Patent Office

Le Président de l'Office européen des brevets
p.o.

Hatten

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Dielectric composition, method of manufacturing a cermic multilayer element and electronic device

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Dielectric composition, method of manufacturing a ceramic multilayer element and electronic device

The invention relates to a dielectric composition comprising a mixture of

- a ceramic composition containing $Ba_aRE_bTi_cO_3$, wherein RE represents a rare earth element, and $0.05 \leq a \leq 0.25$, $0.525 \leq b \leq 0.70$ and $0.85 \leq c \leq 1.0$ and $2a + 3b + 4c = 6$, and being free from lead and bismuth, and
- a glass composition comprising SiO_2.

The invention also relates to a method of manufacturing a ceramic multilayer element comprising the steps of

- manufacturing a multilayer stack comprising a first ceramic foil, a first electrode comprising Cu, a second ceramic foil and a second electrode comprising Cu, which ceramic foils are manufactured from a dielectric composition comprising a ceramic composition and a glass composition comprising SiO_2, which ceramic composition contains $Ba_aRE_bTi_cO_3$, wherein RE represents a rare earth element, and $0.05 \leq a \leq 0.25$, $0.525 \leq b \leq 0.70$ and $0.85 \leq c \leq 1.0$ and $2a + 3b + 4c = 6$, the ceramic composition being free from lead and bismuth; and
- sintering the multilayer stack.

The invention further relates to an electronic device comprising a first dielectric ceramic layer, a first electrode comprising Cu and a second electrode.

Such a dielectric composition is known from EP-A- 0926107. The ceramic composition $Ba_aRE_bTi_cO_3$, wherein RE represents a rare earth element, and $0.05 \leq a \leq 0.25$, $0.525 \leq b \leq 0.70$ and $0.85 \leq c \leq 1.0$ and $2a + 3b + 4c = 6$, is generally known as a group of materials with a high dielectric constant. However, in the case that no glass composition is added, the ceramic composition has a high sintering temperature, generally of more than 1300 °C. It is known that the addition of a glass composition leads to a decrease in the sintering temperature. The known dielectric composition has a sintering temperatures of about 1000°C, which is below 1084°C, the melting temperature of copper. However, as glasses themselves have a low dielectric constant, the composite dielectric composition comprising the ceramic composition and the glass composition has a relatively low dielectric constant as well; the relative dielectric constant is less than 50. In order to get a higher dielectric constant lead or bismuth are generally present in the ceramic composition. Then the

dielectric composition can be sintered at a low temperature. However, due to the presence of lead or bismuth the dielectric composition is toxic and harmful for the environment burned or when set off as waste.

The known dielectric composition further contains CuO. Due to the presence of CuO the known dielectric composition has a higher dielectric constant. The presence of CuO also results in a lower sintering temperature. This CuO is added as powder or as an ingredient of the glass composition. In the case of the use of a Pb-containing ceramic composition, the relative dielectric constant is 62 without the presence of CuO and 77 with the presence of CuO, as follows from the numbered compositions 2,3 and 6 in the cited application.

From the cited application compositions, numbered 21, 22 and 34, are known which are free of lead and bismuth and which can be sintered at a temperature lower than 1100 °C. The glass composition in this dielectric composition comprises 23 % by weight of SiO_2, 14 % by weight of B_2O_3, 61 % by weight of earth alkaline metal oxides and 2 % by weight of Li_2O. The earth alkaline metal oxide comprises 82 % by weight of BaO and at least one species selected from the group consisting of SrO, CaO and MgO. The dielectric compositions referred to further contain CuO. The dielectric compositions have a firing temperature of 1000 °C and a relative dielectric constant of between 27 and 60.

Next to a high dielectric constant it is for electronic applications in the high frequency domain necessary, that the temperature coefficient of the dielectric constant is small. A criterion for this is the NP0-standard, which states that the temperature coefficient of the dielectric constant – also known as the temperature coefficient of capacitance or TCC – should be in the range of –30 to +30 ppm/°C. The cited dielectric compositions that fulfill this criterion and are free of lead and bismuth, have relative dielectric constants between 48 and 53.

Further on, according to this NP0-standard, the dielectric composition should have an RC-time of at least 1000 seconds after sintering. This RC-time has been defined as the product of the resistance against insulation and the capacity, and is a measure of the stability of the dielectric composition.

A disadvantage of said known dielectric compositions that are free of lead and bismuth and of which the TCC fulfills the NP0-standard, is that the relative dielectric constant is relatively low. For miniaturization of devices for high-frequency applications dielectric compositions with high dielectric constants are required.

It is therefore a first object of the invention to provide a dielectric composition of the kind described in the opening paragraph, which composition has a relative dielectric constant higher than 55, of which the TCC fulfills the NP0-standard and which can be sintered with electrodes containing Cu.

It is a second object to provide a method of manufacturing a ceramic multilayer element of the kind described in the opening paragraph, which has a high capacity relative to its surfacial area, of which capacity the temperature coefficient is low.

It is a third object of the invention to provide an electronic device as described in the opening paragraph, which is suitable for high-frequency applications.

The first object is thereby achieved that the dielectric composition contains:

- the ceramic composition,
- as the glass composition a composition comprising SiO_2, a divalent metal oxide chosen from the group consisting of MgO and ZnO and at least 10 weight per cent with respect to the glass composition of a further metal oxide chosen from the group consisting of Li_2O and TiO_2, and

a metal oxide, which is different from the divalent metal oxide that is present in the glass composition.

Surprisingly it has been found that the dielectric composition of the invention fulfills the first object of the invention: a dielectric composition having a relative dielectric constant which is higher than 55, fulfilling the terms of the NP0-standard and which can be sintered together with electrodes containing Cu.

The metal oxide in the dielectric composition of the invention has as a primary function to improve the RC-time. Said RC-time can be calculated as the product of the insulation resistance and the capacity, both of which parameters can be measured. It has turned out that without the presence of a metal oxide the insulation resistance is low after sintering. With such a low insulation resistance of about 10^3 MΩ an RC-time below the NP0-standard is obtained. It has further turned out, that without the presence of a metal oxide the dielectric composition reduces into a semiconducting material during the sintering.

An advantage of the metal oxide is that due to its presence the TCC of the sintered dielectric composition is reduced to values within the limits of the NP0-standard, as follows from the data in table 3. Both effects of the metal oxide – improvement of RC-time and stabilization of TCC - could not be predicted from the presence of CuO in the dielectric compositions of the cited application. According to the cited application, CuO has as a primary function to increase the dielectric constant. Further on, it has turned out that not only

CuO, but also a great variety of other metal oxides can be used in order to improve the RC-time and to stabilize the TCC of the dielectric composition.

Preferably the metal oxide present in the dielectric composition is an oxide of a metal chosen from the group consisting of magnesium, manganese, iron, cobalt, nickel, copper, zinc, yttrium, indium, tungsten, dysprosium, holmium, erbium, ytterbium and lutetium. Table 3 shows the effect on the dielectric constant, the RC-time and the TCC of the presence of a wide variety of metal oxides. All of the preferred metal oxides, except of tungsten oxide, are oxides of so-called "acceptor" metals. The metal ions of these acceptor metals have a charge which is smaller than that of titanium and have a radius which is at most as large as that of titanium. According to the inventors, these metal ions can and partly do replace titanium ions in the ceramic composition, in which the barium rare-earth titanate is generally present in a lattice. In this replacement, also oxygen vacancies are created in the barium rare-earth titanate lattice. Especially in combination with copper electrodes, which brings the condition of a low partial pressure of oxygen, this creation of oxygen vacancies appears to be essential to prevent reduction of the dielectric composition into semiconducting ceramics during sintering. However, the inventors do not want to be bound by said explanation. For example the explanation does not explain, how tungsten oxide acts to prevent the reduction of the dielectric composition during sintering.

Several glass compositions have been found to be effective in lowering the sintering temperature. As the glass composition a variety of silicate glass compositions can be chosen, such as those primarily based on Li_2O-MgO-SiO_2, Li_2O-ZnO-SiO_2, ZnO-SiO_2-TiO_2, MgO-SiO_2-TiO_2, Li_2O-MgO-ZnO-SiO_2-TiO_2. All of these glass composition contain the further metal oxide TiO_2 or Li_2O in an amount of at least 10 weight per cent.

In an embodiment of the dielectric composition of the invention the further metal oxide in the glass composition is Li_2O. As follows from the data in the tables 1 and 2, the dielectric composition with the glass compositions G2, G4, G5, which contain 21, 18 and 14 weight per cent of Li_2O respectively, can be sintered at a lower temperature than the dielectric composition with the glass compositions G1, G3, which contain 2 and 3 weight per cent of Li_2O respectively. However, a man skilled in the art would not use a glass composition containing more than 10 weight per cent of Li_2O. The fact is that it is stated in the cited application that with such a glass composition the dielectric composition has an unsatisfactory moisture resistance after sintering. This unsatisfactory moisture resistance has not been observed by the inventors.

In an embodiment, the glass composition essentially consists of 50-80 weight% of SiO_2, 5-25 weight% of at least one earth alkaline metal oxide, and 10-25 weight% of Li_2O, and is substantially free from boron. With said glass composition good results were obtained. The glass composition has as a further advantage, that it can be used in combination with the binder polyvinyl alcohol, which is water-soluble. In comparison to binders, which are soluble in organic solvents, the water-soluble binder is substantially less harmful to the environment.

Preferably the glass composition is present in an amount of 3 to 5 per cent by weight with respect to the ceramic composition. It has been found that the addition of 3 to 5 per cent by weight of the glass composition of the invention provides a lowering in the sintering temperature of a barium rare-earth titanate ceramic composition of at least 130 °C, from more than 1200 °C to 1070°C or less. This effective lowering of the sintering temperature is huge in comparison with the cited application, in which at least 10 per cent by weight of a glass composition must be added to obtain sufficient sintering at temperatures of about 1000 °C. As a consequence of the small amount of the glass composition present in the dielectric composition, the relative amount of the ceramic composition present in the dielectric composition is large.

In a further embodiment, the earth alkaline metal oxide is primarily MgO. Surprisingly, it has been found that with a glass composition containing MgO and being substantially free from CaO, BaO and SrO the sintering temperature is reduced from 1060 to 1040 °C when using the same amount of glass. As also can be seen in table 3, it appears that the reduction of the sintering temperature takes place while using a lower amount of the glass composition. This results in a higher dielectric constant.

In another embodiment of the dielectric composition of the invention, the divalent metal oxide is ZnO and that the further metal oxide is TiO_2. The glass composition ZnO-SiO_2-TiO_2 is known from EP-A-0960868, which is incorporated herein by reference. In that document no indication is given that this glass composition can be sintered below the melting point of Cu, if a metal oxide different from the divalent metal oxide that is present in the glass composition, is present.

As the ceramic composition, preferably a $Ba_aRE_bTi_cO_3$ is chosen, in which as RE are present Nd and Gd, and in which further the Ba is partially substituted by Sr. Such compositions are known from US-A-5,556,818, which is incorporated herein by reference.

The second object of the invention is thereby achieved, that

- the glass composition contains a divalent metal oxide chosen from the group consisting of MgO and ZnO and at least 10 weight% with respect to the glass composition of a further metal oxide chosen from the group consisting of Li_2O and TiO_2,
- the dielectric composition further contains a metal oxide which is different from the divalent metal oxide that is present in the glass composition, and
- the multilayer stack is sintered at a temperature between 900 and 1080 °C and in an atmosphere which is non-oxidizing for Cu.
- In the method of the invention the dielectric composition of the invention is sintered together with copper electrodes. Examples of atmospheres which are non-oxidising for copper are N_2/H_2O, CO_2/CO and pure N_2. Examples of glass compositions are given in table 1. Also other glass compositions can be used, such as those primarily based on Li_2O-MgO-SiO_2, Li_2O-ZnO-SiO_2, ZnO-SiO_2-TiO_2, MgO-SiO_2-TiO_2, Li_2O-MgO-ZnO-SiO_2-TiO_2. Preferably the sintering takes place at a temperature between 1000 and 1050 °C. Preferably the multilayer stack contains a large number of first and second electrodes, which first and second electrodes are connected to a first and second terminal respectively. The ceramic multilayer element can for example be a ceramic multilayer capacitor, an array of ceramic multilayer capacitors or a high-frequency filter.

The third object of the invention is thereby achieved, in that the first dielectric ceramic layer is a sintered body comprising:

- a ceramic composition containing $Ba_aRE_bTi_cO_3$, wherein RE represents a rare earth element, and $0.05 \leq a \leq 0.25$, $0.525 \leq b \leq 0.70$ and $0.85 \leq c \leq 1.0$ and $2a + 3b + 4c = 6$, and being free from lead and bismuth,
- a glass composition comprising SiO_2, a divalent metal oxide chosen from the group consisting of MgO and ZnO and at least 10 weight per cent with respect to the glass composition of a further metal oxide chosen from the group consisting of Li_2O and TiO_2, and
- a metal oxide, which is different from the divalent metal oxide that is present in the glass composition.

The electronic device of the invention fulfills the object, in that it has a relative dielectric constant of at least 55, that it has a TCC in the range of –30 to +30 ppm/°C and in that it does neither contain lead nor bismuth. As copper has a high electrical conductivity, electrical losses in the electrodes are reduced strongly. Further on, the cost price of the device of the invention is reduced in comparison with a device, in which the electrodes consist of noble metal. Copper can also be present as interconnects, terminals, etcetera, as known in the art of manufacturing electronic devices. As the glass composition a variety of silicate glass

compositions can be chosen, such as those listed in table 1 and those mentioned above. By preference the metal oxide is chosen from the group of consisting of magnesium, manganese, iron, cobalt, nickel, copper, zinc, yttrium, indium, tungsten, dysprosium, holmium, erbium, ytterbium and lutetium. As the ceramic composition, preferably a $Ba_aRE_bTi_cO_3$ is chosen, in which as RE are present Nd and Gd, and in which further the Ba is partially substituted by Sr.

The electronic device may be or may contain a high-frequency filter, such as a filter for electromagnetic interference, an LC- an RC-, an LRC-filter, a common mode filter, a differential mode filter. Alternatively, the electronic device may be a passive component, such as a ceramic multilayer capacitor or an array of capacitors. In a further embodiment, the first ceramic layer, or a stack of ceramic layers with electrodes sandwiched in between of the ceramic layers, is used as a substrate onto which further components are applied by means of depositing and patterning layers or by means of assembling discrete components. The electronic device offers then an integrated solution for a high-frequency application, such as a mobile phone.

Table 1 –glass compositions in weight per cent

	BaO	SrO	MgO	ZnO	Li_2O	TiO_2	SiO_2
G1	26	18	7	0	2	0	47
G2	11	7	3	0	21	0	58
G3	0	0	30	0	3	0	67
G4	0	0	15	0	18	0	67
G5	0	0	9	0	14	0	77
G6	0	0	0	36	0	35	29
G7	0	0	0	28	0	28	44

Table 2 – Sintertemperature T_s and relative dielectric constant K of the dielectric composition comprising a glass and a ceramic composition of $BaGdNdSrTiO_3$, wherein the glass% is the percentage by weight with respect to the ceramic composition

No	glass	glass%	T_s (°C)	K(ε_o)
1*	G1	2	1160	75
2*	G1	5	1130	70
3*	G1	10	1100	60
4*	G1	20	1050	47
5	G2	5	1060	55
6*	G3	5	1100	55
7*	G4	1	1150	65
8	G4	5	1040	55
9	G4	3	1040	62
10	G5	4	1040	55
11**	G6	2	1050	70

* = outside the scope of the present invention

** = further containing 0.5 per cent by weight of CuO

Table 3 - Sintertemperature T_s, RC-time, relative dielectric constant K and temperature coefficient of capacitance TCC of a dielectric composition comprising 4% of the glass G4, a ceramic composition of $BaGdNdSrTiO_3$ and an additive, wherein the additive% is the percentage by weight with respect to the ceramic composition. Sintering took place in an atmosphere which is non-oxidizing for Copper (H_2O/N_2), except for example 2, wherein the partial oxygen pressure was larger.

number	Additive	additive%	T_s (°C)	K (ε_o)	RC-time (10^3 s)	TCC(ppm/°C)
1*	-	0	1040	130	0.0001	very large
2*	-	0	1040	60	0.07	-45
3	CuO	0.5	1052	66	1.3	-18
4	CuO	1.0	1052	65	2.1	-16
5	CuO	2.0	1045	69	3.1	-14
6	MnO_2	0.5	1030	66	8.3	-19
7	MnO_2	0.5	990	62	5.0	-19
8	Co_3O_4	0.5	1050	68	4.5	-7
9	ZnO	0.5	1041	66	4.0	-12
10	Y_2O_3	0.5	1030	68	5.5	-26
11	Y_2O_3	1.0	1041	64	3.0	-30
12	Dy_2O_3	0.5	1048	70	5.1	-2
13	Ho_2O_3	0.5	1048	70	1.7	-4
14	Er_2O_3	0.5	1047	71	3.2	-8
15	In_2O_3	0.5	1050	68	5.8	-10
16	WO_3	0.5	1048	72	6.4	-28

* = outside the scope of the present invention

These and other aspects of the dielectric composition, the glass composition, the method of manufacturing and the electronic device will be further explained with reference to the Figure 1, which schematically shows a cross-section of a first electronic device, a ceramic multilayer element.

Example 1

The ceramic multilayer element 10 of Figure 1 comprises a first ceramic layer 11, a first electrode 21 of Cu, which is in electrical contact with a first terminal 31, a second ceramic layer 12, a second electrode 22 of Cu, which is in contact with a second terminal 32. A number of each of said layers 11, 12 and said electrodes 21, 22 are present in the ceramic multilayer element 10, which results in a multilayer stack. The ceramic multilayer element 10 is a capacitor,

In order to manufacture the ceramic multilayer element 10, a dielectric composition was prepared by mixing 50 grams of $Ba_{0.231}Gd_{0.196}Nd_{0.270}Sr_{0.020}Ca_{0.050}Ti_{1.00}O_3$, 2 grams of a glass composition containing $Mg_{0.18}Li_{0.60}Si_{0.52}O_{1.00}$, 0.25 grams of Y_2O_3, 50 grams of water and a suitable dispergent. After milling in a ball mill with yttrium stabilized zirconia balls for 20 hours an organic binder containing polyvinyl alcohol was added to the dielectric composition. The mixture was subjected to wet mixing to prepare a ceramic slip. This ceramic slip was subjected to sheet molding by the doctor blade method to obtain a rectangular green sheet having a thickness of 21 μm. Next, a conductive paste comprising Cu was printed on the ceramic green sheet 11,12 to form an electrode 21,22. A plural number of the above-mentioned ceramic green sheets 11, 12 to which the electrode 21,22 was formed were laminated, such that between two ceramic green sheets 11, 12 one electrode 21,22 was present.

The resulting structure was dried and subsequently pressed using a pressure of 10 atmosphere. Then the resulting structure was sintered in a kiln, wherein the kiln was heated slowly from room temperature to 1030 °C and subsequently slowly cooled to room temperature. The total firing time was about 9 hours. During the firing a gas flow of N_2/H_2O was used.

After providing the first and the second terminals in the standard way the ceramic multilayer capacitor was ready. The K-value was 68, the loss factor tan δ was 2.10^{-4}. The capacitance was 33 pF. The temperature coefficient of capacitance was – 26 ppm/°C. The insulation resistance was 7.10^{6} MΩ, giving a RC-time of 5500 seconds.

CLAIMS:

1. A dielectric composition comprising a mixture of:

- a ceramic composition containing $Ba_aRE_bTi_cO_3$, wherein RE represents a rare earth element, and $0.05 \leq a \leq 0.25$, $0.525 \leq b \leq 0.70$ and $0.85 \leq c \leq 1.0$ and $2a + 3b + 4c = 6$, and being free from lead and bismuth,
- a glass composition comprising SiO_2, a divalent metal oxide chosen from the group consisting of MgO and ZnO and at least 10 weight per cent with respect to the glass composition of a further metal oxide chosen from the group consisting of Li_2O and TiO_2, and
- a metal oxide, which is different from the divalent metal oxide that is present in the glass composition.

2. A dielectric composition as claimed in Claim 1, characterized in that the metal oxide in the dielectric composition is an oxide of a metal chosen from the group consisting of magnesium, zinc, copper, manganese, cobalt, iron, nickel, erbium, holmium, indium, dysprosium, tungsten and yttrium.

3. A dielectric composition as claimed in Claim 1, characterized in that the further metal oxide in the glass composition is Li_2O.

4. A dielectric composition as claimed in Claim 3, characterized in that the glass composition essentially consists of 50-80 weight% of SiO_2, 5-25 weight% of at least one earth alkaline metal oxide including MgO, and 10-25 weight% of Li_2O and that it is substantially free from boron.

5. A dielectric composition as claimed in Claim 4, characterized in that the earth alkaline metal oxide is primarily MgO.

6. A dielectric composition as claimed in Claim 1, characterized in that in the glass composition the divalent metal oxide is ZnO and that the further metal oxide is TiO_2.

7. A dielectric composition as claimed in Claim 1, 3, 4 or 6, characterized in that the glass composition is present in an amount of 3 to 5 weight per cent with respect to the ceramic composition.

8. A method of manufacturing a ceramic multilayer element comprising the steps of

- manufacturing a multilayer stack comprising a first ceramic foil, a first electrode comprising Cu, a second ceramic foil and a second electrode comprising Cu, which ceramic foils are manufactured from a dielectric composition comprising a ceramic composition and a glass composition comprising SiO_2, which ceramic composition contains $Ba_aRE_bTi_cO_3$, wherein RE represents a rare earth element, and $0.05 \leq a \leq 0.25$, $0.525 \leq b \leq 0.70$ and $0.85 \leq c \leq 1.0$ and $2a + 3b + 4c = 6$, the ceramic composition being free from lead and bismuth; and
- sintering the multilayer stack,

characterized in that

- the glass composition contains a divalent metal oxide chosen from the group consisting of MgO and ZnO and at least 10 weight% with respect to the glass composition of a further metal oxide chosen from the group consisting of Li_2O and TiO_2,
- the dielectric composition further contains a metal oxide which is different from the divalent metal oxide that is present in the glass composition, and
- the multilayer stack is sintered at a temperature between 900 and 1080 °C and in an atmosphere which is non-oxidizing for Cu.

9. An electronic device, comprising a first dielectric ceramic layer, a first electrode comprising Cu and a second electrode, characterized in that the first dielectric ceramic layer is a sintered body comprising:

- a ceramic composition containing $Ba_aRE_bTi_cO_3$, wherein RE represents a rare earth element, and $0.05 \leq a \leq 0.25$, $0.525 \leq b \leq 0.70$ and $0.85 \leq c \leq 1.0$ and $2a + 3b + 4c = 6$, and being free from lead and bismuth,
- a glass composition comprising SiO_2, a divalent metal oxide chosen from the group consisting of MgO and ZnO and at least 10 weight% with respect to the glass composition of a further metal oxide chosen from the group consisting of Li_2O and TiO_2, and

a metal oxide, which is different from the divalent metal oxide that is present in the glass composition.

10. An electronic device as claimed in Claim 9, characterized in that the first dielectric ceramic layer is present as a substrate.

ABSTRACT:

The dielectric composition contains a mixture of a ceramic composition containing $Ba_aRE_bTi_cO_3$, wherein RE represents a rare earth element, and $0.05 \leq a \leq 0.25$, $0.525 \leq b \leq 0.70$ and $0.85 \leq c \leq 1.0$ and $2a + 3b + 4c = 6$, and being free from lead and bismuth, of a glass composition and a metal oxide. The glass composition preferably contains ZnO or MgO, SiO_2 and at least 10 weight% of Li_2O or TiO_2. Preferably, the earth alkaline metal oxide is MgO. By preference, the glass composition essentially consists of 50-80 weight% of SiO_2, 5-25 weight% of MgO and optionally another earth alkaline metal oxide, and 10-25 weight% of Li_2O and is substantially free from boron. The dielectric composition can be sintered in the presence of electrodes of Cu at a temperature below the melting point of Cu to manufacture an electronic device such as a ceramic multilayer element. After sintering the dielectric composition has a relative dielectric constant of at least 55.

Fig. 1

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